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Ferromagnetic–insulator–ferromagnetic tunneling: Spin-dependent tunneling and large magnetoresistance in trilayer junctions (invited)

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Tunneling between ferromagnet–insulator–ferromagnet (FM–I–FM) trilayer thin-film planar junctions has been successfully studied. Tunnel current was observed to be dependent on the relative orientation of the magnetization (M). Co, CoCr, CoFe, $Fe_{0.7}Pt_{0.3}$, and NiFe were tried as the FM electrodes with Al_2O_3 or MgO as the barrier layers for the above studies. Large magnetoresistance (MR) was observed as the M alignment of the two ferromagnets changed from being parallel to antiparallel orientation. At room temperature, the highest change in junction MR was 18%, field sensitivity factor reaching 5%/Oe in the best cases. The MR value increased to 25.6% at 4.2 K, and decreased as the dc bias was increased to a fraction of the barrier height. The angular dependence of MR varied nearly as the cosine of the relative angle of M, as predicted by Slonczewski's theory. The magnitude of MR agrees well with that given by Julliere's model, which predicts that the MR varies as the product of the conduction electron spin polarization of the FMs. These trilayer junctions can find application as high-density, nonvolatile storage media or as field sensors. [S0021-8979(96)50208-5]

INTRODUCTION

Spin-polarized tunneling experiments between a superconductor and a ferromagnet, performed 25 years ago,[1,2] led the way to the present field-dependent tunneling between FM films. Based on the earlier spin-polarized tunneling results, Julliere put forward a model for FM–I–FM tunneling,[3] assuming that spin is conserved in tunneling[2] and tunnel current is dependent on the density of states of the two electrodes. Due to the uneven spin distribution of conduction electrons at the Fermi level in the FMs, one can expect tunneling probability to be dependent on the relative magnetization orientation of the FM films. Analyzing the tunnel current, based on the spin density of states at the Fermi level of the two FM electrodes, Julliere showed the change in the tunnel conductance (ΔG) between antiparallel and parallel orientation of the two FMs as

$$\Delta G/G = 2P_1P_2/(1+P_1P_2), \tag{1}$$

where G is the conductance when the magnetizations are parallel, and P_1 and P_2 are the spin polarizations of the two FM electrodes. The change in the tunnel resistance can be written as

$$\Delta R/R = (R_A - R_P)/R_A = 2P_1P_2/(1+P_1P_2), \tag{2}$$

where R_A and R_P represent the junction resistances when the two FMs have their M antiparallel and parallel, respectively. *This latter definition is used in deducing the MR throughout the present work.*

Another theory of FM–I–FM tunneling proposed by Slonczewski analyzes the transmission of charge and spin currents through a rectangular barrier separating free-electron-like FM metals.[4] The orientation of the spins tunneling across the FM–I interface was strongly influenced by the barrier height, according to this theory. This affected the spin polarization and the exchange coupling between the FMs and was consistent with the low values of $\Delta R/R$ seen previously.[5–11]

The experimental values have come far short of the expected value according to Julliere's model, until very recently. For instance, in the case of Fe–I–Co tunnel junctions, with $P=40\%$ and $P=34\%$, respectively,[1,2] the expected junction MR should be 24%. Julliere studied Fe–Ge–Fe and Fe–Ge–Co junctions and obtained a change of 14% in the conductance at 4.2 K and zero bias.[3] This percentage dropped to less than 1% upon the application of several mV of dc bias to the junction. Various other groups, using mainly NiO, CoO, Gd_2O_3, and Al_2O_3 barriers, studied this effect and only small changes were seen—no more than 7% at 4.2 K.[5–11] In recent times, Miyazaki and his co-workers have improved the room-temperature results from 2.7% to 15.6% [according to the above definition, Eq. (2)].[12] However, the 15.6% value was seen in only one junction and was not reproducible. Using low-temperature deposition techniques, it was recently shown that reproducible results can be obtained using Al_2O_3 barriers with the highest change being 12% for CoFe/Al_2O_3/Co tunnel junctions at room temperature.[13]

Many limiting factors can reduce the MR effect: orange peel coupling between the FMs due to surface roughness,[14] interfacial and barrier spin scattering, FM surface degradation, domain walls, etc. By significantly overcoming some of these negative factors in the present work, values of $\Delta R/R$ have greatly improved to as high as 18% at room temperature and 25.6% at 4.2 K for CoFe/I/Co junctions. This is

close to the expected value of 27.6% predicted by Julliere's model. In the present work, a sensitivity factor of 5%/Oe is obtained for the tunnel junction devices.

EXPERIMENT

The thin-film planar junctions used for MR measurements were fabricated *in situ* through vacuum evaporation, in a system with a background pressure of 10^{-7} Torr. Glass substrates cooled to 77 K were covered with 1 nm thick Si (which serves as a nucleating layer beneath the first electrode). Then, the first FM base electrode, 8.0–15.0 nm thick and 0.2 mm wide, was deposited without or with an applied magnetic field (H) to align with an easy axis of magnetization. The base electrode deposition was followed by the evaporation of 1.2–2.0 nm of Al or Mg metal, covering the *entire* base electrode, to form the tunnel barrier.[15] The substrates were then warmed to room temperature and subjected to oxygen glow discharge to create an insulating layer of Al_2O_3 or MgO. Finally, the top FM electrode, 8.0–25.0 nm thick and 0.3 mm wide, was deposited. The FM materials used include Co, CoCr, CoFe, $Fe_{0.7}Pt_{0.3}$, and NiFe. The thickness was monitored by a quartz-crystal oscillator. In each run, a total of 72 junctions, each with an area of 6×10^{-4} cm^2, were prepared. The junction resistance (R_J) ranged from hundreds of ohms to tens of kilohms.

Junction MR was measured using a four-terminal ac or dc technique in fields up to ±0.5 T, in the temperature range of 4.2–325 K. For ac measurements, a Linear Research-400 ac resistance bridge with an accuracy of ±0.005% was used and dc measurements also had similar accuracy. Measurements were done with H parallel or perpendicular to the junction plane. Several junctions were subjected to higher dc voltages to elucidate the bias dependence of the junction properties. These measurements were taken with voltages ranging from 0 to 0.8 V at 4.2, 77, and 295 K. Detailed temperature dependence of R_J, $\Delta R/R$, and tunnel current was measured between 4.2 and 295 K for some junctions. To control the temperature, the junctions were mounted on a sample probe with a heater. A platinum resistance thermometer, mounted adjacent to the sample, monitored the temperature. Angular dependence was measured at room temperature with the junction plane parallel to the field. R_J was recorded as the sample was rotated from 0° to 360°.

RESULTS

Several criteria were used to qualify the junctions for magnetotransport studies. As a first requirement, junctions with a resistance in the range of 1 kΩ to tens of kilohms were chosen for measurements. To prove that tunneling is the main source of conductance, $G(V)$ and $I(V)$ characteristics were taken for several junctions. Figure 1 shows the conductance of a CoFe/Al_2O_3/Co junction as a function of both low and high voltages at 4.2 K. At low bias, the conductance is nearly constant. However, at high bias nearly a parabolic dependence is observed. This behavior is expected for a good tunnel junction with a barrier height of above ~1 eV. At 4.2 K, a conductance dip was generally seen at zero bias,

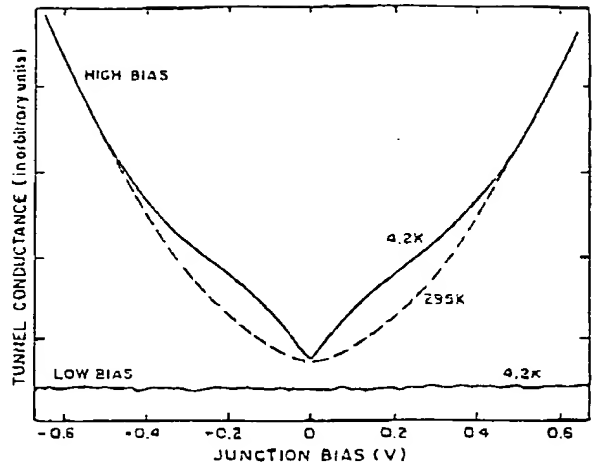

FIG. 1. Tunnel conductance as a function of dc bias for a CoFe/Al_2O_3/Co junction at 4.2 K. The lower curve was taken in the lower bias region and the upper curve is for the higher bias. The voltage scale for the lower curve should be divided by 500. For the higher bias region, data at 295 K are also shown.

whereas at 77 K and room temperature, the conductance dip was negligible. This feature will be discussed later.

$I-V$ curves taken from 0 to 0.8 V also confirm the quality of the junctions. At low bias, ohmic behavior was seen and higher bias showed parabolic dependence. Simmon's theory of tunneling was used to derive the barrier height (ϕ) and thickness (s) for several junctions.[16] For Al_2O_3 barriers, ϕ ranged from 1.8 to 3.5 eV. The corresponding barrier thicknesses varied from 1.8 to 1.2 nm. For the limited studies done with MgO barriers, values of $\phi\cong0.9$ eV and $s\cong2.1$ nm

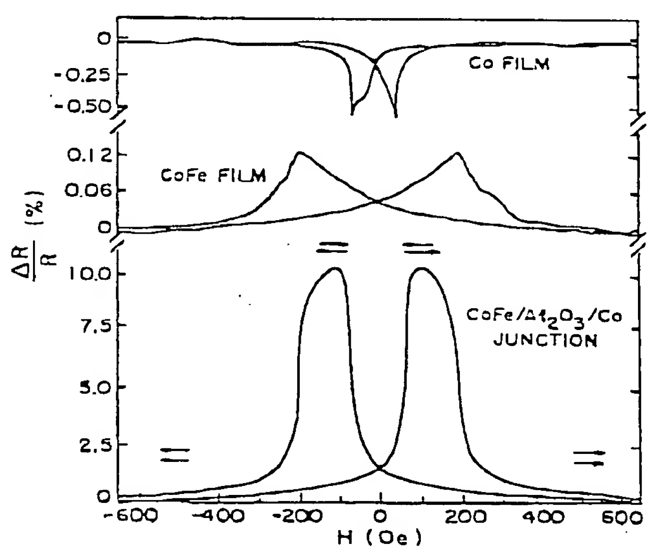

FIG. 2. Magnetoresistance of the tunnel junction and the two FM film electrodes as a function of applied magnetic field, taken at room temperature. The arrows indicate the direction of M in the two FMs, according to the FM-I-FM model of Julliere. (See Discussion).

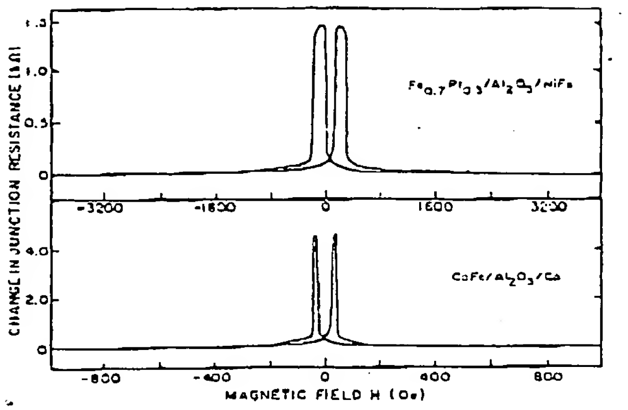

FIG. 3. Junction magnetoresistance vs applied magnetic field showing the variation in the higher field range for $CoFe/Al_2O_3/Co$ and $Fe_{0.7}Pt_{0.3}/Al_2O_3/NiFe$ samples. Note the field independence of MR at fields slightly beyond the peak.

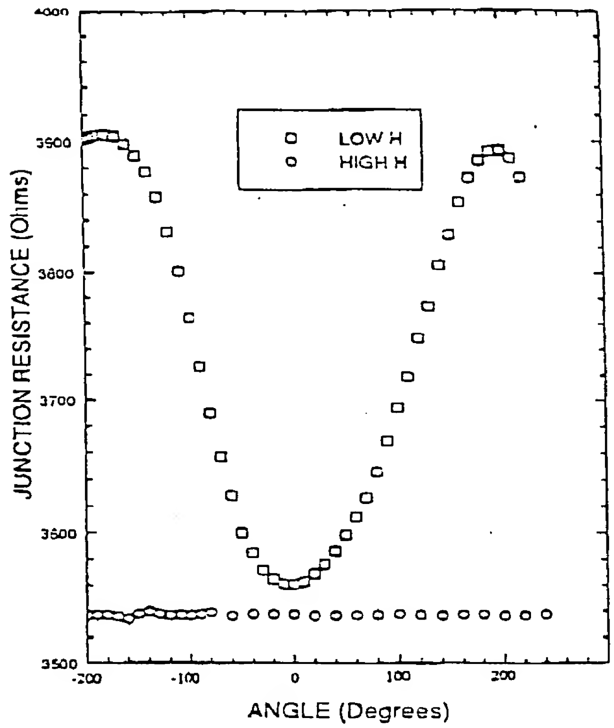

FIG. 4. Angular dependence of the junction magnetoresistance taken at low and high applied fields for a $CoFe/Al_2O_3/Co$ junction.

were obtained. These barrier parameters are what is typically seen for standard Al_2O_3 and MgO tunnel barriers.

Good tunnel junctions with the above barrier parameters showed mostly a small temperature dependence of R_J. The junctions used in the present studies showed only a 10% to 30% increase in R_J as temperature was decreased from room temperature to 4.2 K. According to Simmon's and Stratton's tunneling theory, the tunnel current varies linearly with T^2.[17] This behavior was observed in the present junctions at temperatures <60 K. Also, these junctions withstood temperature cycling, high voltages (up to ±0.8 V), and were stable in the ambient conditions.

MR measurements, displayed in Figs. 2 and 3, show a change in R_J as a function of applied field (H). At high field, the junction resistance is low, and it begins to increase as H decreases toward zero field. Upon reversing the field direction, R_J rapidly rises, showing a peak. With further increase in H,R_J drops to its original low value. *The ratio of the peak change to the absolute value at the peak is defined as* $\Delta R/R$. (This definition gives a lower value of the ratio as compared to the criteria used by others where the resistance change was divided by the low value of R_J at high H). In Fig. 2, the $CoFe/Al_2O_3/Co$ junction shows a $\Delta R/R$ of 10.6% at 300 K. Compared to the junction, the MR of the FM electrodes is also shown. The changes in the resistance for the full length of the strips are about 0.6% and 0.1% for Co and CoFe films, respectively. The junction area corresponds to only 1/30th of the length of the electrodes. In essence, the MR contribution of the FM film is negligible. Figure 3 shows the R_J vs H in the higher field range for two junctions at room temperature. The constancy of R_J as H increased beyond the peak value is evident.

MR effects were investigated with different FM electrodes like Co, CoCr, CoFe, $Fe_{0.7}Pt_{0.3}$, and NiFe and barriers of Al_2O_3 or MgO. Keeping CoFe as one electrode showed the highest percentage change. The maximum change was 25.6% at 4.2 K and 18% at room temperature for the $CoFe/Al_2O_3/Co$ trilayer. Many junctions consistently showed changes in the range of 14%–17% at room temperature. The peak position at room temperature for various junctions occurred between 3 and 120 Oe. For example, $CoFe/Al_2O_3/Co$ and $CoFe/Al_2O_3/NiFe$ junctions deposited in the absence of H showed the peak at ±120 Oe, whereas the peak for junctions prepared in an applied field occurred at ±3 Oe. The peaks occurred at lower field values for thicker NiFe films and for films that were deposited in the presence of an applied field. The peak position and height did not change with field cycling. Also, upon reaching the maximum of R_J, when the field was turned off, the peak value of the resistance was maintained at $H=0$.

The angular dependence of MR was measured for several junctions, for a low and a high value of H, keeping H in the film plane. In order to perform this measurement, the junctions were subjected to a high field in the junction plane in one direction. Upon reversing the field, a low value of H was set (less than the peak field value). R_J was then recorded every 5°–10° as the sample was rotated with respect to the field. Seen in Fig. 4 is the periodic variation of R_J as a function of angle, showing cosine of the angle dependence (not shown in the figure) as discussed in Slonczewski's theory.[4] When similar measurements were done at a field value higher than the peak field, R_J remained constant (the lower curve in the figure). The maximum change in R_J (for low field) with respect to the angle nearly correlated with the $\Delta R/R$ observed in R vs H measurements.

In general, the MR of the junctions increased as the temperature was lowered. For example, in some cases, $\Delta R/R$

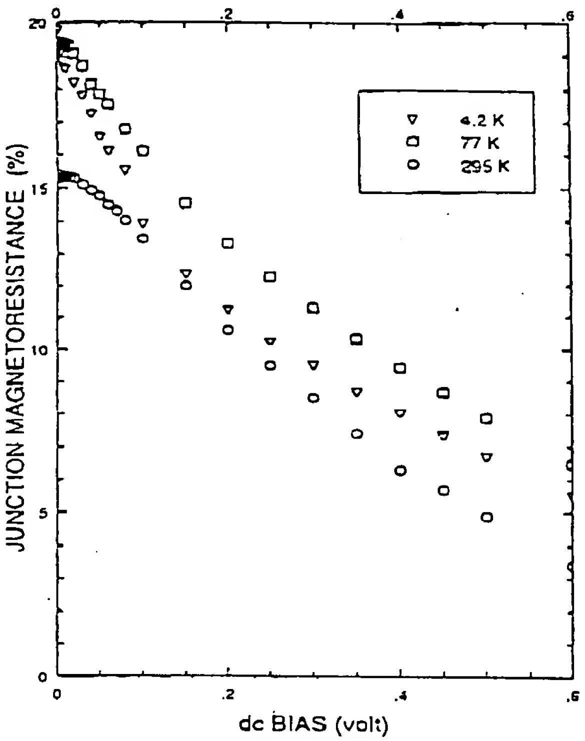

FIG. 5. The dc bias dependence of junction MR for a CoFe/Al_2O_3/Co junction at 4.2, 77, and 295 K.

nearly doubled as the sample was cooled from room temperature to 77 K, and continued to increase as T decreased to 4.2 K. However, in certain cases, the $\Delta R/R$ slightly decreased upon cooling. This will be discussed later. Some junctions were warmed to 325 K and found to be stable. However, the $\Delta R/R$ value slightly decreased at higher temperatures.

The magnetoresistance dependence on the dc bias is displayed in Fig. 5 for a CoFe/Al_2O_3/Co junction. As the dc bias increased, the $\Delta R/R$ increased slightly or remained constant at low bias. At higher voltages, the $\Delta R/R$ declined rapidly, reaching lower MR values. It is important to note that up to a few tens of mV, the decrease is not significant. Junctions with MgO barriers also displayed similar bias dependence of MR. Thicker Al junctions showed lower MR and the visible presence of unoxidized Al. The larger change in the R_J as a function of temperature and considerable asymmetry in the $G(V)$ curve indicated that for thinner barriers (below 1.4 nm), the base electrode was partially oxidized.

DISCUSSION

The junction magnetoresistance can be understood based on Julliere's model.[3] Referring to Fig. 2, at high fields, the two FM films have their M aligned in the applied H direction. (The M direction is indicated by arrows.) Upon reversing the field, the magnetization of the film with a lower coercive force (H_C) aligns itself in the new field direction. On the other hand, the second electrode, with a higher H_C, remains magnetized in the original field direction. In this configuration, the magnetization of the two films are antiparallel to each other. As the field is increased further, it becomes strong enough to align the M of the second FM electrode in the new field direction, resulting in parallel orientation. Thus, at high fields in either direction, the FM electrodes are saturated and parallel to each other. At intermediate fields, the electrodes are aligned antiparallel. According to the model presented in the introduction, when M are parallel, the tunneling probability is highest and tunneling current is maximum, thereby yielding a low R_J. In the antiparallel configuration, the tunneling probability and the current are lowest, resulting in higher R_J.

The MR curves of the two electrodes (see Fig. 2) clearly support the above interpretation. For instance, the extrema points of these curves indicate the coercive forces of the electrodes.[18] The peak in R_J is centered between the lower H_C of Co and the higher H_C of the CoFe film. Thus the peak position of the junction resistance is strongly correlated to the coercive fields of the FM electrodes. The H_C of the FM film is easily influenced by the film growth conditions: the presence of an applied magnetic field during film growth, substrate temperature, a nucleating layer, thickness of the film, etc.[19] CoFe/Al_2O_3/(NiFe or Co) junctions with the films deposited in the presence of an applied field showed a behavior different from those discussed above. For instance, the peak in R_J occurred at 3 Oe as compared to the peak in CoFe/Al_2O_3/(NiFe or Co) junctions, deposited in $H=0$, which occurred at about 120 Oe. Also, the peaks were extremely sharp, with a peak width of less than 3 Oe as compared to over 100 Oe in the other case. The sharpness and low H value of the peak position are directly the result of film growth in the presence of field, which reduces the H_C of the FM film and makes it sharper.[19] Thus, among many other factors, the sharpness and position of the H_C of two FM electrodes strongly influences the MR of the junction.

As pointed out in the previous section, the MR remained constant all the way to 0.5 T, beyond 2 or 3 times the peak value of the applied field (Fig. 3). During the oxidation of Al to form the Al_2O_3 barrier, it is possible that the top surface of the base electrode is partially oxidized as well, creating perhaps an antiferromagnetic (AF) oxide layer like CoO or FeO. In such a situation, one ends up with an FM/AF interface which can exhibit exchange anisotropy, perhaps detrimentally influencing the polarization of the electrons at the interfaces. The presence or absence of exchange anisotropy can be inferred by the shape of R_J vs H at high fields [20] and also by the displaced $M-H$ loop. As seen in Fig. 3, the near absence of MR at fields beyond ~0.05 T, and the symmetric $M-H$ loop observed point to the absence of exchange anisotropy caused by an AF oxide. The data in Fig. 3 were taken at room temperature. However, at 4.2 K, R_J vs H was slightly asymmetric, showing some MR effects even beyond 0.15 T. The $M-H$ loop was also asymmetric at 4.2 K showing the possible presence of an AF layer over the FM.[14]

The rotation of the magnetization of one FM with respect to the other also supports the FM–I–FM tunneling model. This is evident in the data shown for a CoFe/Al_2O_3/Co junction in Fig. 4. At a field value higher

than the H_C of one electrode, when the sample is rotated in a magnetic field, the M of the softer film follows the field. This will change the relative orientation of M in the two FM films—switching from parallel to antiparallel orientation gradually. The tunneling probability is therefore affected, which is evidenced by the periodic variation of the R_J with the angle, as seen in this figure. The maximum value of the resistance corresponds to the antiparallel alignment of the relative magnetizations. The parallel orientation of M goes with the minimum of the curve. At field values higher than the H_C of both electrodes, if the above interpretation is correct, rotation of the sample in the field should maintain the parallel orientation of M (along the applied field) at all angles of H. In this case, the tunneling probability should be the highest and R_J should maintain a constant low value. This is exactly what is seen in the second curve which was taken at high field. Thus the angular dependence and the field dependence of R_J conclusively support the FM–FM tunneling model. The exact form of angular dependence of MR was not quite the cosine function predicted in Slonczewski's theory for the tunnel conductance.[4] The likely distribution of the H_C in the present FM films can contribute to this behavior.

The magnetoresistance behavior of the junction with applied magnetic field in the film plane and perpendicular to it was similar. In perpendicular field, the peaks were relatively broader and occurred at slightly higher field values. The latter peaks were several times broader than the peaks with H in the plane, if the films were deposited in a magnetic field parallel to the substrate plane. However, the MR of the junction remained the same. $M-H$ measurements showed that CoFe and Co films have their M in the film plane. So, when the field is perpendicular to the film plane, the film has to rotate toward the hard axis, thereby shifting the peaks to higher field values and also making them broader; the shift becomes even larger when the films are deposited in an applied magnetic field. Despite these variations, the tunneling model as discussed above is operative here, i.e., even when the magnetizations are perpendicular to the film plane.

Magnetoresistance, junction resistance, tunnel current, and peak position all varied with temperature. As the temperature decreased from 300 to 4.2 K, the MR nearly always increased. The H_C of the elemental FM increased more than that of the alloy FM as the temperature decreased. This allowed broader separation of H_C for the two FMs in some cases, if they were near each other at RT, creating a better antiparallel alignment (wider peaks). Also, at lower temperatures the $M-H$ loop of the FM films got sharper, with a higher value of H_C and remanent magnetization. This can give rise to larger MR effects and peaks shifting to higher H as T decreased. Room-temperature values for $\Delta R/R$ varied between 10% and 18% whereas at 4.2 K, the values went up to 21%–26%. Exceptions to the increase in $\Delta R/R$ with decreasing temperature occurred when the coercive fields of the two films changed such that they moved toward each other with decreasing temperature, creating a sharper peak and bringing down the MR effect. For instance, in the case of certain CoFe/Al_2O_3/NiFe junctions where the FM films were deposited in the presence of H, the H_C were so close to each other that the width of the peak was less than 3 Oe.

The R_J increased with a decrease in the temperature in all cases, the thinner barriers showing a higher percentage change. This is not unexpected since a thinner Al film barrier would allow possible oxidation of the base electrode, thus forming a temperature-dependent, semiconducting barrier at the interface. Such strongly paramagnetic oxide in the interface region can create spin scattering at RT, whereas at lower temperatures, magnetic ordering of this layer would reduce the spin scattering. If this is the case, MR can increase as temperature decreased.

The MR shows surprisingly strong decrease as the bias increased from 0 to 0.8 V. There was no change in $\Delta R/R$ for ac bias up to 1 mV. With dc bias, although $\Delta R/R$ initially showed a small increase or near constant value below ~10 mV at RT, overall it decreased with increasing dc bias. The decrease was small up to about 100 mV, and beyond which, it decreased faster. The value of $\Delta R/R$ at 0.8 V was about 1/5 of the low bias value; MgO barriers showed more decrease as compared to Al_2O_3 barriers. Also, if the $\Delta R/R$ at $V=0$ was lower, then the decrease with bias was larger. Irrespective of the starting $\Delta R/R$, the relative change was similar at 300, 77, and 4.2 K. Many causes can contribute to the decrease in the MR with bias. Among them, the bias dependence of the barrier height, creation of magnons, density of states effects in the FM, etc., may be responsible for the decrease in $\Delta R/R$. With the increase in dc bias, the effective barrier height decreases. MgO, with its smaller barrier height, is affected more than the Al_2O_3 barrier. This can increase the tunneling probability for the minority spins. The spin polarization of the tunneling current can then be expected to decrease, as discussed in Slonczewski's theory.[4] This might also explain strong bias dependence of the effect seen in Julliere's experiment.[3] The a-Ge (although exposed to oxygen) tunnel barrier which Julliere used is known to have a barrier height of 20–30 meV.[21] In this experiment, MR effects were only seen at helium temperatures below about 10 mV. This is the general trend that has been observed with other low barriers like NiO, Gd_2O_3, CoO, etc., where the junction MR effects were seen only at low temperatures and smaller bias.[5-11]

The presence of metal particles,[22] magnons,[23] magnetic impurities, localization effects, multistep tunneling, and states in the barrier or at the interface can adversely affect the spin polarizations of the tunneling electrons by causing spin flip scattering.[24] The dip in the conductance at $V=0$ observed at 4.2 K can be due to some combination of the above causes. One would expect by the presence of metal particles in the tunnel barrier (for example, unoxidized Al left in the barrier) a characteristic $G(0)$ vs T dependence below 4.2 K.[22] No measurable temperature dependence of $G(0)$ was observed between 1 and 4.2 K. However, if the metal particles are less than a few Å, then no significant temperature dependence can be expected.[22] In addition to a conductance dip, small features were observed in G vs V curves around 100 mV, especially in junctions with thinner barriers. This is consistent with the magnon formation as has been found in NiO by Tsui *et al.*,[23] causing spin flip during tunneling. Also, that the anomaly in G at $V=0$ is not the

cause of the MR is evident from the near independence of G vs V and large MR at room temperature, and only a small decrease in the effect at low bias (below $\sim$100 mV), even at low temperatures.

The conduction electron spin polarization values that are used here were measured at the Fermi level. At high bias, one deals with the states away from the Fermi level which need not have the same spin polarization. If such is the case, then at higher bias, the effect ($\Delta R/R$) might decrease if the polarization is lower away from the Fermi level. Spin-polarization measurements at energies higher than at the Fermi level would be helpful. The magnitude of the MR effect can be calculated, for example, for $CoFe/Al_2O_3/Co$ junctions according to Julliere's model [Eq. (2)]. The conduction electron spin polarization of 47% for CoFe was measured at 0.4 K using the spin-polarized tunneling technique as described in Refs. 1 and 2. Using a value of 34% for Co polarization,[2] one gets a calculated change of 27.6%, from Eq. (2). This compared well with the MR seen in the present junctions at 4.2 K.

CONCLUSIONS

Large magnetoresistance was observed in the FM–I–FM tunnel junctions (where FM=Co, CoCr, CoFe, FePt or NiFe, and I=MgO or Al_2O_3). This effect increased at lower temperatures. A significant decrease in MR was observed with increasing dc bias. At room temperature, MR values of 14%–17% were consistently obtained. The highest value of 18% was seen in $CoFe/Al_2O_3/Co$ junctions. This value increased to 25.6% at 4.2 K which agrees well with the value of the MR estimated from the conduction electron spin polarization of the FMs, using Julliere's model. These trilayer devices and MR effects have potential as ultralow power field sensors or memory elements due to the small size, high R_J, independence of FM, barrier layer thickness, and field orientation.

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